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PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In re application of

Docket No: Q79484

SONE, Takuo, et al.

Appln. No.: 10/759,299

Group Art Unit: 1713

Confirmation No.: 7184

Examiner: Kelechi Chidi Egwim

Filed: January 20, 2004

For: METHOD OF PRODUCING CONJUGATED DIENE POLYMERS

**SUBMISSION OF APPEAL BRIEF**

**MAIL STOP APPEAL BRIEF - PATENTS**


Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Submitted herewith please find an Appeal Brief. A check for the statutory fee of \$500.00 is attached. The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account. A duplicate copy of this paper is attached.

Respectfully submitted,

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WASHINGTON OFFICE

**23373**

CUSTOMER NUMBER

Date: March 15, 2006



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**APPEAL BRIEF UNDER 37 C.F.R. § 41.37**

**MAIL STOP APPEAL BRIEF - PATENTS**

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450

Sir:

In accordance with the provisions of 37 C.F.R. § 41.37, Appellant submits the following:

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**I. REAL PARTY IN INTEREST**

The real party in interest is JSR Corporation, the assignee of the present application. The assignment was recorded in the parent application, U.S. Application No. 09/033,685, on March 3, 1998, at Reel 9071, Frame 0445.

## **II. RELATED APPEALS AND INTERFERENCES**

Appellants, Appellants' legal representatives, and the assignee of the present application submit that there was an Appeal in the parent application, U.S. Application No. 09/033,685, Appeal No. 2002-0166. The Notice of Appeal in the parent application was filed on August 8, 2000. The Appeal Brief was filed on February 7, 2001. The Board rendered its decision on the appeal, Appeal No. 2002-0166, on November 18, 2003 (herewith in Appendix). In the decision, the Board upheld the Examiner's rejection of claims 1-4, 8-14 and 19 under 35 U.S.C. § 103 as allegedly being unpatentable over Hattori et al., EP 0267675A2, Ansell et al., WO 93/05083, and Tsujimoto et al., JP 08-073515. In reaching its decision, the Board found that Appellants did not carry their burden of showing unexpected results over Hattori et al., EP 0267675A2, Ansell et al., WO 93/05083, and Tsujimoto et al., JP 08-073515. Additionally, the Board reversed the rejection of claims 1-4, 9-14 and 19 under 35 U.S.C. § 103 as allegedly being unpatentable over Hattori and Tsujimoto since the Board found that the Examiner acknowledged that the catalyst suggested by Hattori and Tsujimoto lacks a halogenated silicon compound. The parent application was abandoned and a continuation application, U.S. Application No. 10/759,299, was filed.

The claims in the parent application, U.S. Application No. 09/033,685 were directed to, "A method of producing a conjugated diene compound with a catalyst consisting essentially of the following components (a) to (d) in an organic solvent; and then reacting with at least one compound selected from the group consisting of the components (e) to (j)...." The Examiner issued an election of species requirement. In response, Applicants elected the halogenated

silicon compound as Component (b), the election of Component (g) from among Components (e)-(j) and the election of Y' = oxygen (epoxy compound) as the specie for Component (g).

The present claims on appeal are different than the claims in the parent application and are directed to, “A method of producing a conjugated diene compound with a catalyst consisting essentially of the following components (a) to (d) in an organic solvent; and then reacting with at least one compound selected from the group consisting of the components (i) to (j). . . .” The present claims do not include Component (g), which was elected during the election of species requirement in the parent application. Additionally, the rejection presented by the Examiner in the present case is an obviousness rejection over Ikematsu et al. , JP 05059103 (“Ikematsu ‘103”) or Ikematsu et al., JP 05051406 (“Ikematsu 406”), in combination with Tsujimoto et al., JP 8073515 (“Tsujimoto”), which is not the same rejection as considered by the Board in the previous Appeal.

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### **III. STATUS OF CLAIMS**

Claims 1-15 and 18-20, all of which are rejected, are the subject of this appeal. Claims 16-17 are canceled. The claims are set forth in their entirety in the Appendix attached to this Brief on Appeal.

#### **IV. STATUS OF AMENDMENTS**

A Response under 37 C.F.R. § 1.116 was filed on October 14, 2005 in response to the Final Office Action dated July 15, 2005. The Examiner indicated in the Advisory Action dated October 28, 2005 that the Amendment would be entered by the Examiner. However, Applicants did not amend the claims in the Response under 37 C.F.R. § 1.116 filed on July 15, 2005. Thus, the claims pending are the claims as amended on April 26, 2005. The Notice of Appeal was filed December 15, 2005.



## **V. SUMMARY OF THE CLAIMED SUBJECT MATTER**

Appellants' invention relates to a method of producing a conjugated diene polymer which comprises polymerizing a conjugated diene compound with a catalyst consisting essentially of components (a) to (d) in an organic hydrocarbon solvent; and then reacting with at least one compound selected from the group consisting of components (i) and (j) (page 4, line 24 to page 5, line 3).

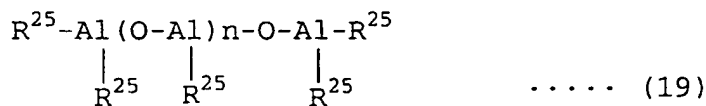
The conjugated diene compound may be selected from the group consisting of 1,3-butadiene, isoprene, 2,3-dimethyl-1,3-butadiene, 1,3-pentadiene, 1,3-hexadiene and cyclo 1,3-hexadiene (page 17, line 28 to page 18, line 8).

Component (a) is a compound containing a rare earth element of atomic number 57-71 in the Periodic Table or a compound obtained by reacting the compound with a Lewis base (page 5, lines 4-7). Component (a) may be selected from the group consisting of a carboxylate, an alkoxide, a  $\beta$ -diketone complex, a phosphate and a phosphite of neodymium, praseodymium, cerium, lanthanum or gadolinium (page 7, line 14 to page 8, line 7).

Component (b) is a compound containing at least one halogen atom (page 5, lines 8-9). Component (b) may be selected from a metal halide, an organometallic halide, an organic halide compound and a halogenated silicon compound (page 10, lines 17-23). The metal halide or organometallic halide may be selected from diethylaluminum chloride, ethylaluminum sesquichloride, ethylaluminum dichloride, diethylaluminum bromide, ethylaluminum sesquibromide and ethylaluminum dibromide (page 10, line 24 to page 11, line 20). The organic halide compound may be selected from benzoyl chloride, xylene dichloride, xylene dibromide,

propionyl chloride, benzyl chloride, benzylidene chloride, t-butyl chloride and the like; organic bromine compounds such as benzoyl bromide, propionyl bromide, benzyl bromide, benzylidene bromide, t-butyl bromide and the like; methyl chloroformate, methylbromoformate, chlorodiphenyl methane and chlorotriphenyl methane (page 11, line 21 to page 12, line 2). The halogenated silicon compound may be selected from trimethylchlorosilane, methyldichlorosilane, diethyl dichlorosilane, methyl trichlorosilane, ethyl trichlorosilane, trichlorosilane, dichlorotetramethyl disilane, dichlorotetramethyl disiloxane and silicon tetrachloride (page 12, line 3 to page 13, line 9).

Component (c) is an aluminoxane (page 5, line 10). Component (c) may have a chemical structure of the following general formula (19) or (20):



wherein  $\text{R}^{25}$  is a hydrocarbon group having a carbon number of 1-20, and  $n$  is an integer of not less than 2 (page 13, line 19 to page 14, line 2). The hydrocarbon group represented by  $\text{R}^{25}$  in the formula (19) or (20) may be a methyl group, ethyl group, propyl group, n-butyl group, isobutyl group or t-butyl group (page 14, lines 3-11).

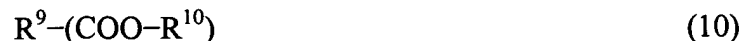
Component (d) is an organoaluminum compound represented by the following general formula (1):



wherein  $R^1$  and  $R^2$  are the same or different and are a hydrocarbon group having a carbon number of 1-10 or a hydrogen atom, and  $R^3$  is a hydrocarbon group having a carbon number of 1-10 provided that  $R^3$  is the same as or different from  $R^1$  or  $R^2$  (page 5, lines 11-18). Component (d) may be selected from the group consisting of trimethylaluminum, triethylaluminum, tri-n-propylaluminum, tri-isopropylaluminum, tri-n-butylaluminum, tri-isobutylaluminum, tripentylaluminum, trihexylaluminum, tricyclohexylaluminum, trioctylaluminum, diethylaluminum hydride, di-n-propylaluminum hydride, di-n-butylaluminum hydride, di-isobutylaluminum hydride, ethylaluminum dihydride, n-propylaluminum dihydride and isobutylaluminum dihydride (page 14, line 24 to page 15, line 15).

Component (a) may be used in an amount of 0.0001-1.0 mmol per 100 g of the conjugated diene compound and the catalyst may have such a composition ratio that a molar ratio of component (a) to component (b) is 1:0.1-1:15, a molar ratio of component (a) to component (c) may be 1:1-1:5000, a molar ratio of component (a) to component (d) may be 1:1-1:500 and a molar ratio of component (c) to component (d) may be 1:0.02-1:300 (page 16, line 3 to page 16, line 16).

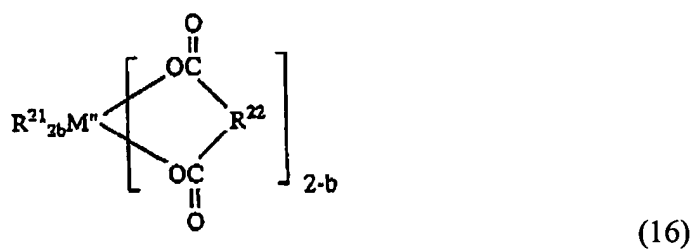
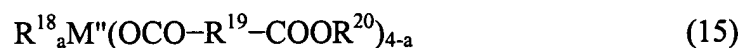
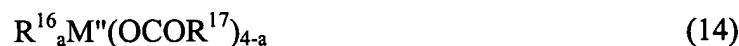
Component (i) is a carboxylic acid, an acid halide, an ester compound, a carboxylic ester compound or an acid anhydride represented by the following general formula (8), (9), (10), (11), (12) or (13):





wherein  $R^7$  to  $R^{15}$  are the same or different and are a hydrocarbon group having a carbon number of 1-50, X is a halogen atom and m is an integer of 1-5 (page 6, line 18 to page 7, line 3); and

Component (j) is a metal salt of a carboxylic acid represented by the following general formula (14), (15) or (16):



wherein  $R^{16}$  to  $R^{22}$  are the same or different and are a hydrocarbon group having a carbon number of 1-20,  $M''$  is a tin atom, a silicon atom or a germanium atom, and a is an integer of 0-3 and b is an integer of 0-1 (page 7, lines 4-12). Component (j) may be selected from the group consisting of dioctyltin dilaurate, dioctyltin bisoctylmaleate, dioctyltin bisbenzylmaleate,

dioctyltin bisethylmaleate and bisoctyltin maleate (page 27, line 21 to page 32, line 24).

Component (j) may be used as a modifying agent (page 32, lines 25-26).

The amount of each of the components (i) and (j) used to the component (a) may be 0.01-200 as a molar ratio (page 32, line 27 to page 33, line 8).

The conjugated diene polymer obtained by using a catalyst consisting essentially of components (a) to (d) may have a content of cis-1,4-bond of not less than 90% and a ratio of weight-average molecular weight to number-average molecular weight of not more than 4 as measured by a gel permeation chromatography (page 19, lines 9-15). The resulting polymer after the polymerization with the catalyst may have content of cis-1, 4-bond of not less than 90% and a ratio of weight-average molecular weight to number-average molecular weight of not more than 3.5 as measured by a gel permeation chromatography (page 19, lines 16-27).

Objects of the present invention include producing conjugated diene polymers where the conjugated diene compound can be polymerized with a high reactivity by using a catalyst system of a rare earth element compound and an aluminoxane and further reacting the resulting polymer just after the polymerization with a specified halide, heterocumulene compound, hetero three-membered-ring containing compound, carboxylic acid, ester compound, carboxylic ester compound, acid anhydride, metal salt of carboxylic acid or the like as a modifying agent to produce a conjugated diene polymer having improved wear resistance, mechanical properties and reduced cold flow (page 4, lines 10-23).

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**VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

The rejection of Claims 1-15 and 18-20 under 35 U.S.C. § 103(a) over Ikematsu et al., JP 05059103 (Ikematsu '103) or Ikematsu et al., JP 05051406 ("Ikematsu '406"), in combination with Tsujimoto et al., JP 8073515, is to be reviewed on appeal.

## **VII. ARGUMENT**

As a preliminary matter, the Examiner has rejected claims 1-15 and 18-20 under 35 U.S.C. § 112, first paragraph, as allegedly failing to comply with the written description requirement.

In the Final Office Action dated July 15, 2005, the Examiner stated that the claim contains subject matter which is not described in the specification in such a way as to reasonably convey to one skilled in the art that the inventor, at the time the application was filed, had possession of the claimed invention. In the Advisory Action dated October 28, 2005, the Examiner acknowledged Applicants' position that the original filed claims contained an error in formula (16), but the Examiner concluded that these errors were not obvious errors, but were different compounds.

On November 17, 2005, in a personal interview, Examiner Egwim indicated that Applicants' arguments were in fact persuasive. Thus, the Examiner agreed to withdraw the rejection of claims 1-15 and 18-20 under 35 U.S.C. § 112, first paragraph, because the new formula in question falls within the scope of the original formula and the embodiments outside the scope of the new formula would not have been viable. See also the Statement of Substance of Interview filed on December 15, 2005.

The Examiner has rejected claims 1-15 and 18-20 under 35 U.S.C. § 103(a) as allegedly being unpatentable over Ikematsu et al., JP 05059103 or JP 05051406 ("Ikematsu '103" and "Ikematsu '406"), in combination with Tsujimoto et al., JP 8073515 ("Tsujimoto '515").

The Examiner's position is that Appellants are arguing against the reference individually and one cannot show non-obviousness by attaching references individually where the rejections are based on combinations of references (page 3 of the Final Office Action dated July 15, 2005).

Contrary to the Examiner's assertion, Appellants are not arguing over the references individually, but instead are arguing that there is no motivation for combining Ikematsu '103 or Ikematsu '406 and Tsujimoto '515. Additionally, Appellants are arguing that even if the references were combined, the present invention provides unexpectedly superior results as compared with the results expected from teaching Ikematsu '103, Ikematsu '406, and Tsujimoto '515.

Ikematsu '103 and Ikematsu '406 teach methods of producing conjugated diene polymers comprising polymerizing a conjugated diene in the presence of a catalyst system in an inert organic solvent and reacting the resulting polymer with a carboxylic acid compound. Ikematsu '103 and Ikematsu '406 teach that the catalyst system comprises a) a lanthanum series (atomic number 57-71), b) an organoaluminum compound and c) a halide compound. However, Ikematsu '103 and Ikematsu '406 do not disclose or suggest that the catalyst consists of an aluminoxane, component (c) in the claimed invention. The Examiner has acknowledged this deficiency in the disclosure of Ikematsu '103 and Ikematsu '406 (page 4 of the Non-final Office Action dated January 26, 2005).

The Examiner asserts that Tsujimoto '515 teaches that the catalyst system consists of an aluminoxane (Non-final Office Action dated January 26, 2005). Further, the Examiner asserts that Tsujimoto '515 discloses using aluminoxane in the catalyst for the purposes of obtaining



narrow molecular weight distributions and higher cis-1,4-bond contents (Non-final Office Action dated January 26, 2005).

Tsujimoto '515 does not disclose or suggest a method of producing a conjugated diene polymer in which a conjugated diene compound is polymerized with a catalyst consisting essentially of (a) to (d) in an organic solvent, and then reacting it with at least one compound selected from the group consisting of components (i) and (j). Since the conjugated diene in Tsujimoto '515 is not reacted with a modifying agent, such as Appellants' components (i) and (j), the polymers in Tsujimoto '515 are weak in reinforcing effect on carbon black and the improving width of the properties is insufficient. As described in Comparative Example 1, 2 and 4 (pages 51 and 57) of Appellants' specification, even if the molecular weight distribution of the polymer itself is small and the content of cis-1,4-bond is high, since the polymer is not modified, the properties of vulcanizate (tan  $\delta$ , wear resistance) are poor as compared with those of Examples 16-18 (page 50) and 26-35 (pages 57 and 61).

Additionally, in the present case, Appellants have provided a § 132 Declaration (herewith in the Evidence Appendix) with experimental data to show that one of ordinary skill in the art would not combine Ikematsu '406 and Tsujimoto '515 and also that the present invention (a combination of the inventions disclosed in Ikematsu '406, and Tsujimoto '515) provides a greater than expected result as compared with the expected results from combining Ikematsu '406 with and Tsujimoto '515.

"A greater than expected result is an evidentiary factor pertinent to the legal conclusion of obviousness...of the claims at issue." *In re Corkill*, 711 F.2d 1496, 226 USPQ 1005 (Fed.

Cir. 1985) (the claimed combination showed an additive result when a diminished result was expected).

In the § 132 Declaration, Run-1 corresponds to Example 11 of the present invention in which the modified polymer is produced by synthesizing a polymer with the catalyst system described in Tsujimoto '515 and subjecting to a coupling reaction with the ester compound described in Ikematsu in order to clearly show the effect of the present invention. This modified polymer has the following features: (1) the molecular weight distribution is broader than that of Tsujimoto '515 by the coupling reaction; (2) the ester compound bonded to the terminal of the polymer is weak in the interaction with the filler.

In Run-1, the modified polymer is produced by synthesizing a polymer with the catalyst system described in Tsujimoto '515 and then is subjected to a coupling reaction with the ester compound described in Ikematsu '406 to show the effects of the present invention. In Run-1, the molecular weight is broader than that of Tsujimoto '515 by the coupling reaction and the ester compound bonded to the terminal of the polymer is weak in its interaction with the filler.

Since the polymer of Run-1 does not have the functional group having an interaction with the filler in the terminal of the polymer, the broad molecular weight distribution would greatly effect the properties which are desirable in the present invention. It would be expected based on what is known in the art that the wear resistance and low heat buildup would be deteriorated in Run-1 as compared with Run-2 because the polymer obtained by the combination of Ikematsu '406 and Tsujimoto '515 has a molecular weight distribution of 3.4, which is broader than the molecular weight distribution of 2.2 in the polymer of Run-2, which corresponds to the

invention of Tsujimoto '515. However, the properties of Run-1 are unexpectedly superior as compared with those of Run-2, which is contrary to the knowledge in the art and it is impossible to predict this effect based the teachings of Ikematsu '406 and Tsujimoto '515.

Additionally, Appellants submit that even if the coupling reaction of Ikematsu '103 (modification with a carboxylic acid or acid anhydride after the polymerization with a lanthanide catalyst) was used in place of the coupling reaction disclosed in Ikematsu '406 (modification with an ester compound after the polymerization with a lanthanide catalyst), the results of combining Ikematsu '103 and Tsujimoto '515 would be the same as the result produced when combining Ikematsu '406 and Tsujimoto '515.

The Examiner asserts that Appellants' § 132 Declaration only demonstrates possible additional advantages which would flow naturally from following the suggestion of the prior art (Tsujimoto '515) to include the aluminoxane catalyst component (c) in the catalyst mixture of components a, b and d, in order to reduce the molecular weight distribution of the diene polymer while increasing the molecular weight (see Tables 1, 3 and 6 of Tsujimoto '515) and the suggestion in the prior art (Ikematsu '103 and Ikematsu '406) to after treat the resulting diene polymer resulting from such catalytic polymerizations with the carboxylic acid compound in order to have lower solution viscosities (see abstracts of Ikematsu '103 and Ikematsu '406) (page 4 of the Final Office Action dated July 15, 2005).

Appellants respectfully disagree with the Examiner's assertion that the advantages of the present invention flow naturally Ikematsu '103 or Ikematsu '406 and Tsujimoto '515. As demonstrated in the § 132 Declaration, the advantages of the present invention are unexpected as

compared with the results expected from combining Ikematsu '406 and Tsujimoto '515. When an Applicant demonstrates substantially improved results and states that the results were unexpected, this should suffice to establish unexpected results in the absence of evidence to the contrary. *In re Soni*, 54 F.3d 746, 34 USPQ2d 1684, 1688 (Fed. Cir. 1995). The Examiner has not provided any reasonable basis to question Appellants' comparative data and showing that the superior results are unexpected. The Examiner's position appears to be that he has established a *prima facie* case of obviousness and that after that any unexpected results due to the combination are expected merely because he considers the art is combinable.

Based on extensive experimentation on the relationship between the catalyst system and the polymerization behavior, the inventors have accomplished the present invention. That is, it has been found that the catalyst system according to the present invention has a higher living property and is suitable for the coupling reaction and that the low molecular weight components can be further reduced by utilizing the characteristic of the catalyst system to conduct the coupling reaction. Even in the polymer having a broad molecular weight distribution of Run-1, the excellent vulcanization properties are developed and the cold flow is improved.

For the above reasons, it is respectfully submitted that Appellants' claims would not have been obvious over the combination of Ikematsu '103 or Ikematsu '406 in view of Tsujimoto '515 and the rejection under 35 U.S.C. §103(a) is improper and should be overturned.

Unless a check is submitted herewith for the fee required under 37 C.F.R. §41.37(a) and 1.17(c), please charge said fee to Deposit Account No. 19-4880.

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The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

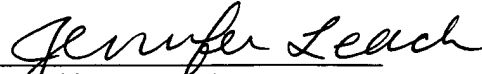
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Date: March 15, 2006

### **VIII. CLAIMS APPENDIX**

#### CLAIMS 1-15 and 18-20 ON APPEAL:

1 (rejected): A method of producing a conjugated diene polymer which comprises polymerizing a conjugated diene compound with a catalyst consisting essentially of the following components (a) to (d) in an organic hydrocarbon solvent; and then reacting with at least one compound selected from the group consisting of the following components (i) and (j).

Component (a) : a compound containing a rare earth element of atomic number 57-71 in the Periodic Table or a compound obtained by reacting the compound with a Lewis base;

Component (b) : a compound containing at least one halogen atom;

Component (c) : an aluminoxane;

Component (d) : an organoaluminum compound represented by the following general formula (1):



wherein  $\text{R}^1$  and  $\text{R}^2$  are the same or different and are a hydrocarbon group having a carbon number of 1-10 or a hydrogen atom, and  $\text{R}^3$  is a hydrocarbon group having a carbon number of 1-10 provided that  $\text{R}^3$  is the same as or different from  $\text{R}^1$  or  $\text{R}^2$ ;

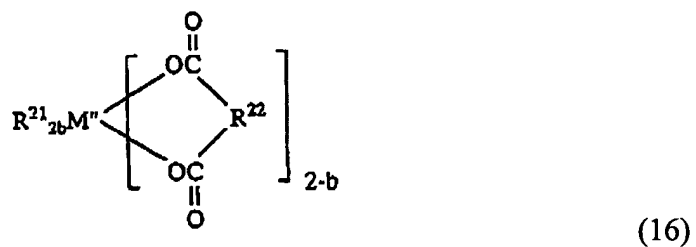
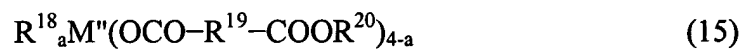
Component (i) : a carboxylic acid, an acid halide, an ester compound, a carboxylic ester compound or an acid anhydride represented by the following general formula (8), (9), (10), (11), (12) or (13):





wherein  $R^7$  to  $R^{15}$  are the same or different and are a hydrocarbon group having a carbon number of 1-50, X is a halogen atom and m is an integer of 1-5; and

Component (j) : a metal salt of a carboxylic acid represented by the following general formula (14), (15) or (16):



wherein  $R^{16}$  to  $R^{22}$  are the same or different and are a hydrocarbon group having a carbon number of 1-20, M" is a tin atom, a silicon atom or a germanium atom, and a is an integer of 0-3 and b is an integer of 0-1.

2 (rejected): The method according to claim 1, wherein the resulting polymer after the polymerization with the catalyst has a content of cis-1, 4-bond of not less than 90% and a ratio of weight-average molecular weight to number-average molecular weight of not more than 3.5 as measured by a gel permeation chromatography.

3 (rejected): The method according to claim 1, wherein the resulting final polymer has a content of cis-1, 4-bond of not less than 90% and a ratio of weight-average molecular weight to number-average molecular weight of not more than 4 as measured by a gel permeation chromatography.

4 (rejected): The method according to claim 1, wherein the component (a) is selected from the group consisting of a carboxylate, an alkoxide, a  $\beta$ -diketone complex, a phosphate and a phosphite of neodymium, praseodymium, cerium, lanthanum or gadolinium.

5 (rejected): The method according to claim 1, wherein the component (b) is selected from a metal halide, an organometallic halide, an organic halide compound and a halogenated silicon compound.

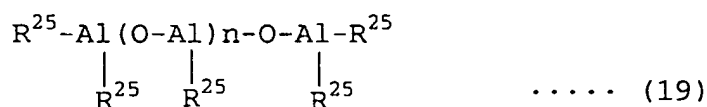
6 (rejected): The method according to claim 5, wherein said metal halide or organometallic halide is selected from diethylaluminum chloride, ethylaluminum sesquichloride, ethylaluminum dichloride, diethylaluminum bromide, ethylaluminum sesquibromide and ethylaluminum dibromide.



7 (rejected): The method according to claim 5, wherein said organic halide compound is selected from benzoyl chloride, xylene dichloride, xylene dibromide, propionyl chloride, benzyl chloride, benzylidene chloride, t-butyl chloride and the like; organic bromine compounds such as benzoyl bromide, propionyl bromide, benzyl bromide, benzylidene bromide, t-butyl bromide and the like; methyl chloroformate, methylbromoformate, chlorodiphenyl methane and chlorotriphenyl methane.

8 (rejected): The method according to claim 5, wherein said halogenated silicon compound is selected from trimethylchlorosilane, methyldichlorosilane, diethyl dichlorosilane, methyl trichlorosilane, ethyl trichlorosilane, trichlorosilane, dichlorotetramethyl disilane, dichlorotetramethyl disiloxane and silicon tetrachloride.

9 (rejected): The method according to claim 1, wherein the component (c) has a chemical structure of the following general formula (19) or (20):



wherein  $\text{R}^{25}$  is a hydrocarbon group having a carbon number of 1-20, and  $n$  is an integer of not less than 2.

10 (rejected): The method according to claim 9, wherein the hydrocarbon group represented by  $\text{R}^{25}$  in the formula (19) or (20) is methyl group, ethyl group, propyl group, n-butyl group, isobutyl group or t-butyl group.

11 (rejected): The method according to claim 1, wherein the component (d) is selected from the group consisting of trimethylaluminum, triethylaluminum, tri-n-propylaluminum, tri-isopropylaluminum, tri-n-butylaluminum, tri-isobutylaluminum, tripentylaluminum, trihexylaluminum, tricyclohexylaluminum, trioctylaluminum, diethylaluminum hydride, di-n-propylaluminum hydride, di-n-butylaluminum hydride, di-isobutylaluminum hydride, ethylaluminum dihydride, n-propylaluminum dihydride and isobutylaluminum dihydride.

12 (rejected): The method according to claim 1, wherein the component (a) is used in an amount of 0.0001-1.0 mmol per 100 g of the conjugated diene compound and the catalyst has such a composition ratio that a molar ratio of component (a) to component (b) is 1:0.1-1:15, a molar ratio of component (a) to component (c) is 1:1-1:5000, a molar ratio of component (a) to component (d) is 1:1-1:500 and a molar ratio of component (c) to component (d) is 1:0.02-1:300.

13 (rejected): The method according to claim 1, wherein the conjugated diene compound is selected from the group consisting of 1,3-butadiene, isoprene, 2,3-dimethyl-1,3-butadiene, 1,3-pentadiene, 1,3-hexadiene and cyclo 1,3-hexadiene.

14 (rejected): The method according to claim 1, wherein an amount of each of the components (i) and (j) used to the component (a) is 0.01-200 as a molar ratio.

15 (rejected): The method according to claim 1, wherein the component (j) is used as a modifying agent.

16 (canceled).

17 (canceled).

18 (original): The method according to claim 15, wherein the component (j) is selected from the group consisting of dioctyltin dilaurate, dioctyltin bisoctylmaleate, dioctyltin bisbenzylmaleate, dioctyltin bisethylmaleate and bisoctyltin maleate.

19 (original): A conjugated diene polymer produced by the method of claim 1 and having a content of cis-1,4-bond of not less than 90% and a ratio of weight-average molecular weight to number-average molecular weight of not more than 4 as measured by a gel permeation chromatography.

20 (rejected): A conjugated diene polymer produced by the method of claim 1, wherein said at least one compound selected from the group consisting of the following components (i) and (j) is component (j).

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**IX. EVIDENCE APPENDIX:**

Pursuant to 37 C.F.R. § 41.37(c)(1)(ix), submitted herewith is a copy of the § 132 Declaration submitted on April 26, 2005, which was entered by the Examiner in the Final Office action dated July 15, 2005, and which is relied upon by Appellant in the appeal.

Appln. No.: 10/759,299  
Appeal Brief under 37 C.F.R. § 41.37

**X. RELATED PROCEEDINGS APPENDIX**

A copy of the Board's decision rendered in Appeal No. 2002-0166 filed in Application No. 09/033,685 is attached herewith.



PP

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opinion in support of the decision being entered today was not written for publication and is not binding precedent of the Board.

Paper No. 30

UNITED STATES PATENT AND TRADEMARK OFFICE

**DOCKETED**

NOV 19 2003

BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

**MAILED**

NOV 18 2003

Ex parte TAKUO SONE, KATSUTOSHI NONAKA,  
IWAKAZU HATTORI and AKIO TAKASHIMA

PAT. & T.M. OFFICE  
BOARD OF PATENT APPEALS  
AND INTERFERENCES

Appeal No. 2002-0166  
Application No. 09/033,685

HEARD: February 4, 2003

Before KIMLIN, PAK, and MOORE, Administrative Patent Judges.  
PAK, Administrative Patent Judge.

DECISION ON APPEAL

This is a decision on an appeal under 35 U.S.C. § 134 from the examiner's refusal to allow claims 1 through 5, 8 through 14 and 19. Claims 6, 7 and 15 through 18, the remaining claims in the instant application, stand withdrawn from consideration by the examiner as being directed to a non-elected invention.

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APPEALED SUBJECT MATTER

According to the examiner (the first Office action dated October 4, 1999 (Paper No. 4, page 4), appellants were

required under 35 U.S.C. [§] 121 to elect a single disclosed species for prosecution on the merits to which the claims shall be restricted if no generic claim is finally held to be allowable.

Pursuant to such a requirement, the appellants elected and prosecuted one of the species recited in claims 1 through 5, 8 through 14 and 19 on appeal. See, e.g., the Reply Brief dated May 17, 2001 (Paper No. 21), pages 4 and 5. Therefore, for purposes of this appeal, we limit our discussion and decision to the elected and prosecuted species recited in the claims on appeal only.<sup>1</sup> See *Ex parte Ohsaka*, 2 USPQ2d 1461, 1461-62 (Bd. Pat. App. & Int. 1987). Both the examiner and the appellants defined the elected and prosecuted species recited in the appealed claims as follows<sup>2</sup>:

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<sup>1</sup> We take no position on the non-elected species covered by the claims on appeal as they have not been examined or rejected by the examiner.

<sup>2</sup> See the Supplemental Examiner's Answer dated March 12, 2003 (Paper No. 26), page 3, and Appellants' Response to the Supplemental Examiner's Answer and to the Remand and Order under 37 CFR 1.196(a) and (d) dated April 11, 2003 (Paper No. 27) (hereinafter referred to as "Supplemental Reply Brief"), page 2.

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A method of producing a conjugated diene polymer which comprises polymerizing a conjugated diene compound with a catalyst consisting essentially of the following components (a) to (d) in an organic hydrocarbon solvent; and then reacting with at least one compound selected from the group consisting of the following component (g):

Component (a): a compound containing a neodymium compound or a compound obtained by reacting the neodymium compound with a Lewis base;

Component (b): a halogenated silicon compound;

Component (c): methylaluminoxane;

Component (d): an organoaluminum compound represented by the following general formula (1):



(wherein  $\text{R}^1$  and  $\text{R}^2$  are the same or different and are a hydrocarbon group having a carbon number of 1-10 or a hydrogen atom, and  $\text{R}^3$  is a hydrocarbon group having a carbon number of 1-10 provided that  $\text{R}^3$  is the same as or different from  $\text{R}^1$  or  $\text{R}^2$ );

Component (g): a hereto three-membered-ring containing compound having a chemical structure of the following general (7):



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(wherein Y' is an oxygen atom).

#### PRIOR ART

As evidence of obviousness, the examiner relies on the following prior art references:

Hattori et al (Hattori)	0 267 675 A2	May 18, 1988
(Published European Patent Application)		
Ansell et al. (Ansell)	WO 93/05083	Mar. 18, 1993
(Published International Patent Application)		
Tsujimoto et al. (Tsujimoto)	08-073515	Mar. 19, 1996 <sup>3</sup>
(Published Japanese Patent Application)		

#### REJECTIONS

The appealed claims stand rejected as follows:

- (1) Claims 1 through 4, 9 through 14 and 19 under 35 U.S.C. § 103 as unpatentable over the combined disclosures of Hattori and Tsujimoto; and
- (2) Claims 1 through 5, 8 through 14 and 19 under 35 U.S.C.

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<sup>3</sup> Our reference to Tsujimoto is to its corresponding English translation of record.

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§ 103 as unpatentable over the combined disclosures of Hattori, Tsujimoto and Ansell.

#### OPINION

We have carefully reviewed the claims, specification and applied prior art references, including all of the arguments and evidence advanced by both the examiner and the appellants in support of their respective positions. This review leads us to conclude that only the examiner's Section 103 rejection of claims 1 through 5, 8 through 14 and 19 as unpatentable over the combined disclosures of Hattori, Tsujimoto and Ansell is well founded. Accordingly, we affirm only this Section 103 rejection for essentially those reasons set forth in the Answer and below.

The examiner finds (Answer, pages 3 and 4), and the appellants do not dispute (Brief, Reply Brief and Supplemental Reply Brief in their entirety), that Hattori teaches a process for producing a modified conjugated diene polymer, comprising polymerizing a conjugated diene in an inert solvent in the presence of a catalyst and then reacting (modifying) the resulting conjugated diene polymer with an epoxide (three-membered heterocyclic ring) containing compound. See also, e.g., Hattori, abstract and page 3, line 32 to page 4, line 4. The examiner finds (Answer, page 4), and the appellants do not dispute (Brief, Reply Brief and

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Supplemental Reply Brief in their entirety), that the catalyst described in Hattori contains a lanthanum series rare earth metal compound (neodymium compound) corresponding to the claimed component (a), an organoaluminum compound corresponding to the claimed component (d), and a halide compound. The examiner finds (Answer, page 4), and the appellants do not dispute (Brief, Reply Brief and Supplemental Reply Brief in their entirety), that Hattori teaches that its process produces polymers with high cis-1, 4 conjugated diene contents of more than 90%. Indeed, Hattori exemplifies processes by which polymers having cis-1,4 conjugated diene contents ranging from 96.8% to 97.3% are produced. See pages 12, Table 1, page 17, Table 2 and page 19, Table 3. Implicit in Hattori's teachings is that these high cis-1,4 conjugated diene contents and modifying agent are important in improving wear resistance and mechanical characteristics. See, e.g., Hattori, page 3, lines 1-8, page 7, line 58 to page 8, line 3.

The examiner recognizes that the catalyst described in Hattori does not contain the claimed methylaluminoxane and the claimed halogenated silicon compound. To remedy these deficiencies, the examiner relies on the disclosures of Tsujimoto and Ansell. The examiner finds (Answer, pages 4 and 6) and the appellants do not

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dispute (Brief, Reply Brief and Supplemental Reply Brief in their entirety), that

[I]t is known in the art to add an aluminoxane to a catalyst system comprising a) a lanthanum series rare earth metal compound, b) an organoaluminum and c) a halide compound for polymerizing conjugated dienes, for the purpose of obtaining even **narrower molecular weight distributions** and **higher cis-1,4-bond contents**, such as taught by Tsujimoto et al. (See ¶ 25 in Full translation of Tsujimoto et al.)

. . . .

[I]t is known in the art to use a halogenated silicon compound as a halide component in a catalyst system for polymerizing conjugated dienes, for the purpose of obtaining products of narrower molecular weight distribution and/or smaller high molecular weight fractions whilst retaining desired very high cis-1,4 bond content and catalyst activity, such as taught by Ansell et al. See page 3, lines 11-15.

The examiner then concludes that one of ordinary skill in the art would have been led to include the aluminoxane described in Tsujimoto and the halogenated silicon compound described in Ansell (as Hattori's halide component) in the polymerization catalyst described in Hattori, motivated by a reasonable expectation of successfully obtaining the advantages taught by Tsujimoto and Ansell.

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The appellants do not dispute the examiner's determination that it would have been *prima facie* obvious to employ the halogenated silicon compound described in Ansell as the halide component of the polymerization catalyst described in Hattori. Compare the Answer, pages 4 and 5, with the Brief, Reply Brief, Supplemental Reply Brief, and Appellants' Response to Examiner's "Office Communication" Dated September 4, 2003 in their entirety. Rather, the appellants appear to argue that one of ordinary skill in the art would not have been led to include the aluminoxane described in Tsujimoto in the polymerization catalyst of Hattori. See the Brief, pages 11 and 12. We do not agree.

The applied prior art references, like the appellants, recognize the importance of obtaining polymers having not only high cis-1,4 conjugated diene polymer contents, but also narrow molecular weight distributions (Mw/Mn). The narrow molecular weight distributions (Mw/Mn) and desired molecular weights, for example, are known to be important "since they affect the processing characteristics of the diene polymer, such as the processability and carbon black incorporation time, the physical properties of the cured polymer ...". See, e.g., Ansell, page 1, line 27 to page 2, line 15. According to Tsujimoto, its improved polymerization catalyst is useful for obtaining conjugate diene

polymers having a high cis-1, 4 structure and a narrow molecular weight distribution from a conjugate diolefin or conjugate diolefin mixture. See pages 1 and 10. We find that Tsujimoto then shows in its examples that the addition of methylaluminoxane to the polymerization catalyst of the type described in Hattori, i.e., a catalyst containing a lanthanum series rare earth metal compound (neodymium compound), an organoaluminum compound, and a halide compound, assists in obtaining conjugate diene polymers having narrower molecular weight distributions ( $M_w/M_n$ ) than those produced by the polymerization catalyst of the type described in Hattori (without methyl aluminoxane as part of the catalyst). See, e.g., pages 20 and 22, Tables 1, 2 and 6, together with pages 14 and 15. Specifically, we find that Tsujimoto teaches that the presence of methylaluminoxane in the polymerization catalyst of the type described in Hattori is useful for producing conjugate diene polymers having narrow molecular weight distributions ( $M_w/M_n$ ) ranging from 2.3 to 3.2 at the polymerization temperature taught by Hattori. Compare Hattori, page 6, lines 7-8 relating to its polymerization temperature with Tsujimoto, pages 13 and 15, relating to its polymerization temperature and pages 20 and 22, Tables 1, 2 and 6, relating to the actual molecular weight distributions. Thus, we concur with the examiner that one of

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ordinary skill in the art would have been led to include methylaluminoxane in the polymerization catalyst of Hattori, motivated by a reasonable expectation of successfully improving the polymer produced by Hattori's process.

As a rebuttal to the *prima facie* case of obviousness established by the examiner, the appellants argue that the showing in the specification and Rule 132 declarations<sup>4</sup> executed by Takuo Sone (one of the inventors in the present application) on June 14, 2000 and August 15, 2000, respectively, demonstrates that the elected subject matter of the appealed claims imparts unexpected results. See the Brief, pages 13-23, the Reply Brief, pages 3-5 and Appellants' Response to Examiner's "Office Communication" Dated September 4, 2003, pages 4-6. However, a close examination of the showing in the specification and Rule 132 declarations reveals that the appellants have not carried their burden of showing unexpected results.

Initially, we find that none of the Runs, experiments or examples in the specification and the Rule 132 declarations, except

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<sup>4</sup> While the first Rule 132 declaration was executed by Takuo Sone (one of the inventors in the present application) on June 14, 2000, the second Rule 132 declaration, otherwise known as the "supplemental declaration" was executed by Takuo Sone on August 15, 2000.

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for Example 24 in the specification and Run No. 9 in the second Rule 132 declaration (referred to by the appellants as the "supplemental declaration"), employs catalytic components and a modifying agent, which are within the elected subject matter of the appealed claims. We find that Example 6 in the specification and Run No. 13 in the second Rule 132 declaration, for example, do not employ catalysts within the elected subject matter of the appealed claims. See the specification, pages 37, 39, 40 and 48 and the second Rule 132 declaration, page 4. The catalysts employed therein are simply devoid of the claimed and elected halogenated silicon compound. *Id.* Similarly, we find that Runs 1 and 5 shown in the first Rule 132 declaration employ neither a catalyst nor a modifying agent within the elected subject matter of the appealed claims.

We find that the appellants have not demonstrated that the conjugated diene polymers produced in Example 24 and Run 9 have unexpected properties. See *In re Merck & Co.*, 800 F.2d 1091, 1099, 231 USPQ 375, 381 (Fed. Cir. 1986); *In re Freeman*, 474 F.2d 1318, 1324, 177 USPQ 139, 143 (CCPA 1973). We find that Example 24 and Run 9 show the production of conjugated diene polymer having a cis-1, 4-bond content of 96.8% and a molecular weight distribution (Mw/Mn) of 2.6. This polymer has a rebound resilience of 67% and a



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wear resistance index of 127. However, as indicated *supra*, the presence of methylaluminoxane in the polymerization catalyst of the type described Hattori is reasonably expected to produce conjugated diene polymer having a cis-1, 4-bond content of 96.8% to 97.3% and a molecular weight distribution of 2.3 to 3.2, which according to the applied prior art references, improve, *inter alia*, wear resistance and other physical characteristics. Moreover, as found by the examiner at page 10 of the Answer and as indicated *supra*, Hattori teaches that wear resistance and other mechanical properties of the conjugated diene polymer can be further improved by modifying it with a particular modifying agent. Thus, we concur with the examiner that the properties referred to in the specification and the Rule 132 declarations are reasonably expected by one of ordinary skill in the art from the teachings of the applied prior art references. See, e.g., *In re Skoner*, 517 F.2d 947, 950, 186 USPQ 80, 82 (CCPA 1975) ("Expected beneficial results are evidence of obviousness of a claimed invention just as unexpected beneficial results are evidence of unobviousness"). The appellants have not proffered "any evidence to suggest that the effects of the aluminoxane taught by Tsujimoto ... and the modifying agent taught by Hattori ... are not simply accumulative

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with regard to [the claimed] properties such as wear resistance."  
See the Answer, page 10.

To the extent that Example 24 and Run 9 evince that the appellants have discovered new additional benefits, it is our judgement that on balance, the expected advantages of employing the polymerization catalyst and modifying agent as suggested by the applied prior art references outweigh the appellants' newly discovered benefits for the reasons well articulated by the examiner at pages 9-11 of the Answer. See *In re May*, 574 F.2d 1082, 1092, 197 USPQ 601, 609 (CCPA 1978); *In re Nolan*, 553 F.2d 1261, 1267, 193 USPQ 641, 645 (CCPA 1977).

Also, we find that the appellants have not demonstrated that the showing in Example 24 and Run 9 is reasonably commensurate in scope with the elected subject matter of the appealed claims. See *In re Clemens*, 622 F.2d 1029, 1035, 206 USPQ 289, 296 (CCPA 1980). While Example 24 and Run 9 are directed to polymerizing a specific diene with a specifically prepared and aged polymerization catalyst containing, e.g., particular amounts of specific neodymium and halogenated silicon **compounds**, under a particular polymerization reaction condition and modifying the resulting polymer with a specific epoxide containing **compound** (styrene oxide), the elected subject matter of the appealed claims is not so limited. Compare

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the elected subject matter of the appealed claims with the specification, pages 53 and 54 and the second Rule 132 declaration, pages 1-3. The elected subject matter of the appealed claims embraces polymerization catalysts, polymerization conditions and epoxide containing compounds (modifying agents), which are materially different from those employed in Example 24 and Run 9.

Moreover, we find that the appellants have not compared the elected subject matter of the appealed claims with the closest embodiment of the closest prior art, i.e., Hattori. *In re Baxter Travenol Labs*, 952 F.2d 388, 392, 21 USPQ2d 1281, 1285 (Fed. Cir. 1991). While Runs 10 and 14 employ a catalyst and a modifying agent within those disclosed in Hattori, they are not directed to a polymerization process for producing conjugated diene polymer having a cis-1, 4 bond content of, e.g., 97.3%, which is the closest to those polymers produced in Example 24 and Run 9.

Finally, we cannot ascertain from the showing in the specification and Rule 132 declarations whether the allegedly improved properties are due to the presence of different amounts of organoaluminum compound, and/or methylaluminoxane as alleged. *In re Dunn*, 349 F.2d 433, 439, 146 USPQ 479, 483 (CCPA 1965) ("While we do not intend to slight the alleged improvements, we do not feel it an unreasonable burden on appellants to require comparative

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examples relied on for non-obviousness to be truly comparative. The cause and effect sought to be proven is lost here in the welter of unfixed variables." ). In this regard, we note that Example 24 and Run 9 employ a different amount of a specific organoaluminum compound in their catalysts than those employed in Runs 10 and 14.

Thus, after due consideration of all of the evidence and arguments proffered by both the examiner and the appellants, we determine that the preponderance of evidence weighs in favor of obviousness. Accordingly, we affirm the examiner's decision rejecting all of the appealed claims under 35 U.S.C. § 103 as unpatentable over the combined teachings of Hattori, Tsujimoto and Ansell.

However, we reverse the examiner's rejection of claims 1 through 4, 9 through 14 and 19 under 35 U.S.C. § 103 as unpatentable over the combined teachings of Hattori and Tsujimoto since the examiner acknowledges that the catalyst suggested by Hattori and Tsujimoto lacks a halogenated silicon compound (elected subject matter). See the Answer, page 6.

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CONCLUSION

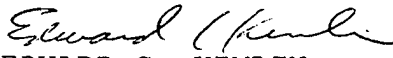
In summary:

- 1) The rejection of claims 1 through 4, 8 through 14 and 19 under 35 U.S.C. § 103 as unpatentable over the combined teachings of Hattori, Tsujimoto and Ansell is affirmed;
- (2) The rejection of claims 1 through 4, 9 through 14 and 19 under 35 U.S.C. § 103 as unpatentable over the combined teachings of Hattori and Tsujimoto is reversed.

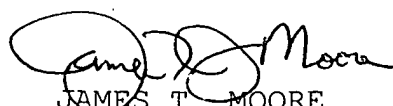
Accordingly, the decision of the examiner is affirmed.

No time period for taking any subsequent action in connection with this appeal may be extended under 37 CFR § 1.136(a).

AFFIRMED

  
EDWARD C. KIMLIN  
Administrative Patent Judge

  
CHUNG K. PAK  
Administrative Patent Judge

  
JAMES T. MOORE  
Administrative Patent Judge

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Appeal No. 2002-0166  
Application No. 09/033,685

SUGHRUE MION ZINN  
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Takuo SONE et al

Serial No. 10/759,299

Group Art Unit: 1713

Filed: January 20, 2004

For : METHOD OF PRODUCING CONJUGATED DIENE POLYMERS

DECLARATION UNDER RULE 132

I, Takuo Sone, declare that:

I am an inventor of the above-referenced United States Patent Application Serial No. 10/759,299.

I received my Doctor of Material Bioengineering from Tokyo University of Agriculture and Technology in the year of 1995 and I have been employed by JSR Corporation since 1995, wherein I have been engaging mainly in the research and development of various solution-polymerized elastomers.

I have made the following experiment in order to show that the present invention can not be easily conceived from Ikematsu (JP 05051406) or Tsujimoto (JP 8073515) or a combination thereof.

Experiment

Five polymers of Run-1 to Run-5 were prepared in the same manner as described in Example 1 of the specification of the present invention using a catalyst system and a modifying agent shown in Table 1. In this case, Run-1 corresponds to Example 11 of the specification of the present invention using as a modifying agent diethyl adipate described in Ikematsu, and Run-2 and Run-3 correspond to Comparative Examples 1

and 2 of the specification of the present invention, which are supplementary experiments of Tsujimoto, and Run-4 is a supplementary experiment corresponding to Comparative Example 1 of Ikematsu and Run-5 is a supplementary experiment corresponding to Example 3 of Ikematsu. In Run-4 and Run-5, the amounts of the catalyst components added were adjusted so as to match the Mooney viscosity with those of Run-1 to Run-3, and the amount of diethyl adipate added was adjusted so as to be equal to that of Example 11.

A vulcanizate was prepared by using each of the polymers of Run-1 to Run-5 according to the compounding recipe described on page 36 of the specification of the present invention in the same manner as described on page 36, lines 11 to 16 of the specification of the present invention.

The properties of the polymer and the vulcanizate were measured in the same manner as described on pages 35 to 36 of the specification of the present invention to obtain results as shown in Table 2.



Table 1

	Catalyst component (mmol)				Modifying agent (mmol)
	Rare earth element compound	Aluminoxane	Organoaluminium compound	Component (b)	
Run-1 (Example-11)	Nd(Oct) <sub>3</sub> 0.18	MAO 18.5	Al <sup>i</sup> Bu <sub>2</sub> H 3.9	AlEt <sub>2</sub> Cl 0.37	Diethyl adipate 3.6
Run-2 (Comparative Example 1)	Nd(Oct) <sub>3</sub> 0.18	MAO 18.5	Al <sup>i</sup> Bu <sub>2</sub> H 3.9	AlEt <sub>2</sub> Cl 0.37	-
Run-3 (Comparative Example 2)	Nd(Oct) <sub>3</sub> 0.18	MAO 4.7	Al <sup>i</sup> Bu <sub>2</sub> H 3.8	AlEt <sub>2</sub> Cl 0.37	-
Run-4	Nd(Oct) <sub>3</sub> 0.21	-	Al <sup>i</sup> Bu <sub>2</sub> H 5.6	Al <sub>2</sub> Et <sub>3</sub> Cl <sub>3</sub> 0.42	-
Run-5	Nd(Oct) <sub>3</sub> 0.21	-	Al <sup>i</sup> Bu <sub>2</sub> H 5.6	Al <sub>2</sub> Et <sub>3</sub> Cl <sub>3</sub> 0.42	Diethyl adipate 3.6

Table 2

	Mooney viscosity ( $ML_{1+4, 100^{\circ}C}$ )	Mw/Mn	Cis-1,4- bond (%)	Vinyl-1,4- bond (%)	Cold Flow (mg/min)	TB (MPa)	EB (%)	Rebound resilience (25°C, %)	Wear resistance (index)
Run-1 (Example-11)	38	3.4	96.9	1.2	1.6	26.5	475	65	123
Run-2 (Comparative Example 1)	31	2.2	97	1.1	18.5	26.5	470	62	120
Run-3 (Comparative Example 2)	34	3.6	97	1.1	15.5	25.8	510	56	105
Run-4	34	3.8	95.8	1.2	6.8	26.1	490	52	94
Run-5	36	4.4	95.8	1.2	4.6	26	490	50	89
Run-6 (Comparative Example 3)	-	-	-	-	-	25.7	530	54	100

In Table 2, Run-6 corresponds to Comparative Example 3 of the specification of the present invention and is a standard sample for evaluating the properties of the vulcanizate.

As seen from the comparison of Run-1 with Run-2 and Run-3, the cold flow is considerably improved by conducting the modification reaction with diethyl adipate.

Then, the rebound resilience and the wear resistance are reviewed as the properties of the vulcanizate. In general, it is known that both the properties are dependent upon the molecular weight distribution of the polymer. Also, both the properties are dependent upon the molecular weight of the polymer. They are deteriorated as the molecular weight of the polymer becomes low. The polymer having a wide molecular weight distribution is unfitted for the improvement of the rebound resilience and wear resistance because it has a greater amount of low molecular weight component. Therefore, the rebound resilience and wear resistance become good in the polymer having a narrow molecular weight distribution if the Mooney viscosity is the same level. In fact, as seen from the comparison between Run-2 and Run-3 corresponding to the examples of Tsujimoto, Run-2 having a narrower molecular weight distribution is excellent in the rebound resilience and wear resistance as compared with Run-3.

In Run-1 corresponding to Example 11 of the present invention, the molecular weight distribution ( $M_w/M_n$ ) is 3.4, which is approximately equal to that of Run-3, but the rebound resilience and wear resistance are excellent as compared with those of Run-2. Since the polymerization conditions in Run-1 are the same as in Run-2, the molecular weight distribution of the polymer before the addition of diethyl adipate is as narrow as 2.2 and also the low molecular weight component is less, and hence when such a polymer is subjected to the

modification reaction with diethyl adipate, the reaction proceeds in a higher efficiency and the low molecular weight component is further decreased and as a result, the rebound resilience and wear resistance are improved. From this fact it is clear that the catalyst system defined in the present invention has a high living property and is very suitable for the modification reaction in a higher reaction efficiency and brings about a high balance the cold flow and the vulcanization properties by the combination with the modification reaction.

Run-4 and Run-5 are the supplementary experiments of Ikematsu as mentioned above. The polymer of Run-4 before the reaction with diethyl adipate has a molecular weight distribution of 3.8 and the polymer of Run-5 obtained by reacting the polymer of Run-4 with diethyl adipate has a molecular weight distribution of 4.4, from which it is clear that the modification reaction with diethyl adipate proceeds. However, the cold flow is 6.8 in Run-4 and 4.6 in Run-5, so that the improving effect of the cold flow is small as compared with Run-1. Also, the rebound resilience and wear resistance in Run-5 are 50 and 89, which are deteriorated as compared with those of Run-4 (52 and 94). This is due to the fact that MAO (methylaluminoxane) is not used in the catalyst system of Ikematsu and the living ratio is low. That is, it is guessed that since the living ratio in the catalyst system of Ikematsu is low, the contribution ratio of low molecular weight component to the modification reaction is low, and hence the cold flow and the rebound resilience and wear resistance are deteriorated due to the influence of widening the molecular weight distribution to 4.4. In the invention of Ikematsu, therefore, it is not always attained that the cold flow and the vulcanization properties are largely improved by simply conducting the modification reaction.

It can be seen from the above experiment that the present invention can not be easily conceived from Ikematsu or Tsujimoto or a combination thereof.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: March 30, 2005      Declarant: Takuo Sone  
Takuo Sone

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